Nuclear spin effects in singly negatively charged InP quantum dots

Bipul Pal,[1,*] Sergey Yu. Verbin,[2,3,†] Ivan V. Ignatiev,[2,3] Michio Ikezawa,[1] and Yasuaki Masumoto[1]

[1]*Institute of Physics and Tsukuba Advanced Research Alliance (TARA), University of Tsukuba, Tsukuba 305-8571, Japan*

[2]*Institute of Physics, St.-Petersburg State University, St.-Petersburg, 198504, Russia*

[3]*Venture Business Laboratory, University of Tsukuba, Tsukuba 305-8571, Japan*

(Dated: February 6, 2008)

Experimental investigation of nuclear spin effects on the electron spin polarization in singly negatively charged InP quantum dots is reported. Pump-probe photoluminescence measurements of electron spin relaxation in the microsecond timescale are used to estimate the time-period T_N of the Larmor precession of nuclear spins in the hyperfine field of electrons. We find T_N to be ~ 1 μs at $T \approx 5$ K, under the vanishing external magnetic field. From the time-integrated measurements of electron spin polarization as a function of a longitudinally applied magnetic field at $T \approx 5$ K, we find that the Overhauser field appearing due to the dynamic nuclear polarization increases linearly with the excitation power, though its magnitude remains smaller than 10 mT up to the highest excitation power (50 mW) used in these experiments. The effective magnetic field of the frozen fluctuations of nuclear spins is found to be 15 mT, independent of the excitation power.

PACS numbers: 78.67.Hc, 71.35.Pq, 72.25.Fe, 72.25.Rb, 71.70.Jp

I. INTRODUCTION

Strong localization of electrons in quantum dots (QDs) may enhance the hyperfine interaction of electron spins with those of nuclei.[1] Various aspects of the hyperfine interaction of electron and nuclear spins have been studied for last three decades in different materials,[2] including InP QDs.[3,4] Charge-tunable InP QDs with one resident electron per dot, on an average, have recently attracted considerable research interest due to the observation of submillisecond spin lifetime of resident electrons in these QDs.[5,6] This observation makes it a promising candidate for quantum memory element in the emerging fields of quantum information technology and spintronics.[7] Application of QDs as the building blocks of quantum computers has been proposed.[8]

However, the influence of the hyperfine interaction between electron and nuclear spins, on the long-lived electron spin polarization needs to be clarified. Two effects of the electron-nuclear spin-spin interactions may be considered. One of them is the so-called dynamic nuclear polarization.[2] In the optical orientation of electron spins in semiconductors by circularly polarized photons, the spin-oriented electrons dynamically polarize the nuclear spins due to the hyperfine coupling of the electron and nuclear spin subsystems.[2,9] In turn, the spin-polarized nuclei produce an effective internal magnetic field (the Overhauser field B_N), which may influence the electron spin dynamics. In presence of an external magnetic field $B_{\rm ext}$, electron spins should feel a total magnetic field $B_T = B_{\rm ext} + B_N$.

Another important effect of the electron-nuclear hyperfine coupling is the electron spin relaxation via its interaction with nuclear spins.[10,11,12] At low temperature, relaxation of the coupled electron-nuclear spin system is determined by three processes, namely, i) electron spin precession in the frozen fluctuations of the hyperfine field of the nuclear spins, ii) nuclear spin precession in the hyperfine field of the electron spins, and iii) nuclear spin relaxation in the dipole-dipole field of its nuclear neighbors. These three processes have very different characteristic timescales. Theoretical estimate for GaAs QDs containing $\sim 10^5$ nuclei suggest them to be ~ 1 ns, ~ 1 μs, and ~ 100 μs, respectively.[10] The last one of the above processes will not be considered further in this paper, because it affects the electron spin dynamics over a long timescale of 10^{-4}–10^{-3} s. In this long time-regime many other mechanisms, such as those originating from the spin-orbit coupling and interaction with phonons become important for electron spin relaxation.

The first relaxation process mentioned above arises from the fact that due to a large, but limited number of nuclear spins, typically $n \sim 10^5$, interacting with the electron spin in a QD, random correlation of nuclear spins may create a fluctuating nuclear polarization, $\Delta F_N \propto F_N/\sqrt{n}$, where F_N is the total spin of the polarized nuclei. Fluctuation ΔF_N acts on the electron spin as another internal magnetic field B_f, with random magnitude and orientation over the QD ensemble.[10] We may note that the Larmor precession of the nuclear spins in the hyperfine field of an electron spin is much slower than that of the electron spin in the nuclear hyperfine field, because the interaction of an electron spin with a single nucleus is $\sqrt{n}$ times weaker compared to its interaction with the effective magnetic field of the nuclear fluctuations. Thus, the electron "sees" a snapshot of the "frozen fluctuations" of the nuclear field. Electron spin precession in the magnetic field B_f is expected to cause electron spin relaxation in the QD ensemble in a timescale of the order of 1 ns, during which the electron spin polarization decays to one-third of its initial value. After the initial decay in the nanosecond timescale, the remaining spin polarization relaxes very slowly over a timescale much longer than the radiative lifetime of the electron-

hole pair.[10]

Electron spin relaxation due to the frozen fluctuations of nuclear spins (FFNS) may be suppressed by a longitudinally (along the optical excitation axis) applied magnetic field with a magnitude larger than B_f.[10] Recent study of electron spin relaxation in p-doped InAs QDs by Braun *et al.*[13] found that at zero external magnetic field, electron spin polarization decays down to one-third of its initial value within 800 ps after photo-excitation. Then the residual spin polarization remains stable with no measurable decay within the photoluminescence (PL) decay time. The authors found that the initial fast relaxation of electron spin was suppressed by the application of a small (~ 100 mT) external magnetic field. We want to note the fact that for $B_{\text{ext}} = 0$, *two distinctly different time regimes are present* in the electron spin relaxation.[14] The initial fast relaxation is caused by the random distribution of B_f in the QD ensemble.[10,13] However, *a total depolarization of electron spins does not take place* in this regime. The residual electron spin polarization decays very slowly in a second relaxation regime, which is governed by the slow time-varying change in the distribution of B_f. This slow change in the distribution of B_f is caused by the variation in the precession rate of nuclear spins in the hyperfine field of electrons.[10,13] We denote it as the nuclear spin precession effect (NSPE). Study of electron spin relaxation due to the NSPE in the microsecond time-range should give an estimate of the nuclear spin precession period T_N in the hyperfine field of electrons. However, to the best of our knowledge, this has not been experimentally studied so far.

In this paper we describe our experimental study of nuclear spin effects on the long-lived spin polarization of resident electrons, observed recently[5,6] in the singly negatively charged InP QDs. Electron spin dynamics and the influence of nuclear spins on it, are probed by the time-resolved as well as time-integrated measurements of the degree of PL circular polarization ρ_c, defined quantitatively in Sec. II. Our time-domain measurements of ρ_c by using a PL pump-probe technique[5,6,15,16] in the microsecond time-regime reveal electron spin relaxation via the NSPE at the vanishing external magnetic field. From the value of the electron spin decay time τ_d for $B_{\text{ext}} = 0$, we estimate that $T_N \sim 1$ μs at $T \approx 5$ K, comparable to the theoretical estimate available for GaAs QDs.[10] We also measure the dependence of ρ_c on B_{ext} in time-integrated measurements at $T \approx 5$ K. From these steady-state measurements we obtain the Overhauser field $B_N = 6$ mT at 50 mW CW excitation and the effective magnetic field of the FFNS, $B_f = 15$ mT, independent of the excitation power. The relatively small value of B_N may come from efficient leakage of QD nuclear spin polarization to the surrounding lattice nuclei.

II. EXPERIMENTAL

Our sample consists of a single layer of self-assembled InP QDs, embedded between $Ga_{0.5}In_{0.5}P$ barriers grown on a n^+-GaAs substrate. The average base diameter (height) of the QDs is ~ 40 (5) nm and the areal density of dots is $\sim 10^{10}$ cm^{-2}. Semi-transparent indium-tin-oxide electrode is deposited on top of the sample to control the charge state of the QDs by means of an applied electric bias.[5,6,17] For the present study on the singly negatively charged QDs we apply an electric bias of $U_b = -0.1$ V. This is because it was found from a previous study of trionic quantum beats[17] on the same sample that at $U_b \approx -0.1$ V the QDs contain one resident electron per dot, on an average.

Electron spins in the QD ensemble are polarized in our experiments by using the well-known optical orientation technique.[2,18] We excite the QDs *quasiresonantly* (in the excited state of the QDs, but below the wetting layer bandgap) by circularly polarized beam from a Ti:Sapphire laser, which can be operated either in CW or in pulsed mode. The excitation beam is directed along the sample growth axis and is focused to a spotsize of ~ 150 μm in diameter on the sample kept in a magneto-optical cryostat at $T \approx 5$ K. The excitation energy $E_x = 1.77$ eV and the detection energy $E_d = 1.72$ eV used in our experiments are indicated by arrows on the polarization-resolved PL spectra in Fig. 1(inset). These spectra are measured by using suitable combinations of retardation plates and Glan-Thompson linear polarizers and a triple spectrometer (focal length 1 m) equipped with a CCD detector. The spectral resolution is 0.05 meV. We monitor the degree of circular polarization $\rho_c = (I_S - I_O)/(I_S + I_O)$ for the ground state PL. Here I_S (I_O) is the intensity of the PL component having the same (opposite) circular polarization as that of the excitation beam. We study ρ_c, in the time-integrated as well as time-resolved measurements, as a function of the external magnetic field B_{ext} applied along the optical excitation axis (longitudinal magnetic field, Faraday geometry). Time-resolved data are taken by using a synchroscan streak camera, while for the time-integrated measurements, a GaAs photomultiplier tube and a two channel gated photon counter are used.

III. RESULTS AND DISCUSSION

A. Negative circularly polarized PL

Our measurements of polarization-resolved PL spectra under quasiresonant excitation ($E_x = 1.77$ eV) of singly negatively charged InP QDs show that the degree of circular polarization ρ_c is *negative*[5,6,19,20] in the spectral region 1.7–1.735 eV, for which $\Delta E = (E_x - E_d)$ lies between 70–35 meV [Fig. 1(inset)]. We measure the time dependence of ρ_c for $E_d = 1.72$ eV. The data is plotted in Fig. 1. Initially ρ_c is seen to be positive, but it becomes

negative at 70 ps after the excitation pulse and then ρ_c approaches a *constant negative value*. We denote this constant value as the amplitude of circular polarization of PL ($A_{\rm CP}$) [see Fig. 1]. The value of $A_{\rm CP}$ increases logarithmically with the excitation power[24] and reaches up to 45%. In the time-integrated measurements negative value of ρ_c reaches up to 30%.

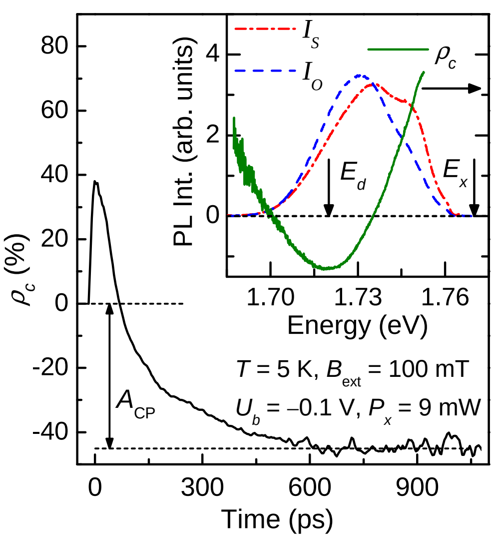

FIG. 1: (Color online) Time-dependence of ρ_c measured for E_x and E_d indicated in the inset. $B_{\rm ext}$ = 100 mT was applied to suppress the effect of the FFNS. The inset shows the spectra of I_S and I_O. Spectral dependence of ρ_c is shown for E_x indicated by an arrow.

The remarkable *negative circular polarization of PL* is related to the optically created spin orientation of the resident electrons. How the sign of ρ_c is determined by the spin direction of the resident electron in a QD is discussed in many papers, see e.g., Refs. 5,6,15,21,25,26. Here we briefly explain how the negative degree of circular polarization of PL arises for our experimental condition, due to the presence of the optically polarized resident electron spins in the QDs. For this, we refer to the schematic diagram of Fig. 2. Quasiresonant excitation in our experiments creates electron-hole pair in the QD excited state. As the resident electron spins in the QDs are polarized by the excitation photons,[18] the spin of the photogenerated electron in the excited state and that of the resident electron in the ground state should have a parallel orientation in the majority of the QDs in the ensemble. For simplicity, we consider here only these QDs [Fig. 2(i)], as ρ_c for the QD ensemble is mainly determined by them. In these QDs, a direct energy relaxation of the photogenerated electron to the ground state is blocked due to Pauli exclusion principle. However, the electron-hole pair in the excited state can undergo a flip-flop transition,[6,21,22] in which a simultaneous flip of the electron and hole spins takes place [Fig. 2(ii)]. This is followed by energy relaxation of both the hot carriers [Fig. 2(iii)]. The flip-flop transition is caused by the anisotropic exchange interaction in QDs.[21,25,26] After the flip-flop transition, PL emitted from the radiative recombination of the spin-flipped electron and hole in the ground state has the opposite circular polarization compared to the excitation photons [Fig. 2(iv)]. Thus, the degree of PL circular polarization becomes negative. We assume that the hole spin relaxation time in the QD ground state is much longer than the radiative lifetime.[27,28] This is supported by the data of Fig. 1, where ρ_c approaches a constant negative value and remains stable over the PL decay time.

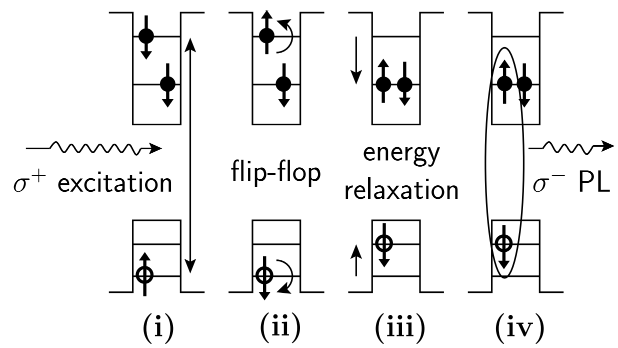

FIG. 2: A schematic model to explain the occurrence of the negative value of ρ_c due to the presence of the optically oriented resident electron spins in the QDs.

The measurements of ρ_c in the time-integrated experiments and of $A_{\rm CP}$ in the time-resolved experiments are used in this work for the study of nuclear spin effects on the spin polarization of resident electrons in the singly negatively charged QDs.

B. Frozen fluctuations of nuclear hyperfine field

It turns out that the PL circular polarization, and hence, the electron spin polarization, is very sensitive to $B_{\rm ext}$. Time-integrated measurements in Fig. 3 show the dependence of ρ_c on $B_{\rm ext}$ for σ^+- and σ^--polarized CW excitations. As seen there, ρ_c becomes nearly independent of $B_{\rm ext}$ for $B_{\rm ext} > 50$ mT. Absolute value of ρ_c decreases with decreasing $|B_{\rm ext}|$ and reaches a minimum for $|B_{\rm ext}|$ nearly, but not exactly zero. The behavior of ρ_c as a function of $B_{\rm ext}$ can be fitted well by a Lorentzian with a half width at half maximum of 15 mT.

The decrease of $|\rho_c|$ may be interpreted as the effect of electron spin relaxation in the QD ensemble by the field B_f of the FFNS.[10] The effect is suppressed by the external magnetic field when $B_{\rm ext}$ exceeds B_f in magnitude, allowing electron spin polarization, and hence, $|\rho_c|$ to increase to reach a steady-state value. So, the value of B_f may be estimated from the half width at half maximum of the Lorentzians in Fig. 3. Thus, we estimate a value of B_f = 15 mT for the QDs under study. The obtained value of B_f is found to be independent of the excitation power up to the highest excitation power (50 mW) used, suggesting that it is intrinsic to the InP

QDs. For $B_f = 15$ mT, we estimate the electron spin relaxation time $\tau_s = \hbar/(g_e \mu_B B_f) \approx 0.5$ ns (μ_B = Bohr magneton and g_e = electron Landé g-factor = 1.5[29]), resulting from the FFNS. The values of $B_f = 15$ mT and $\tau_s = 0.5$ ns obtained here for the InP QDs, are comparable to those estimated theoretically by Merkulov *et al.*[10] for GaAs QDs, and to those obtained experimentally by Braun *et al.*[13] for InAs QDs.

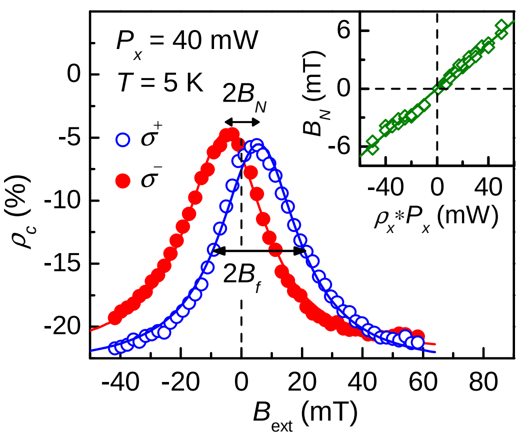

FIG. 3: (Color online) Dependence of ρ_c on B_{ext} for σ^+- and σ^--polarized CW excitations. The solid lines are Lorentzian fits used to estimate B_f. Shifts of the minima of $|\rho_c|$ from $B_{\text{ext}} = 0$ estimate B_N. The inset shows linear dependence of B_N on the scaled excitation power $\rho_x{*}P_x$, where $\rho_x = +1$ (-1) for σ^+ (σ^-) excitation and P_x = excitation power.

C. Dynamic nuclear polarization

As seen in Fig. 3, the minima of $|\rho_c|$ is shifted from $B_{\text{ext}} = 0$. This is caused by the Overhauser field B_N, due to which the electron spin "feels" a total magnetic field $B_T = B_N + B_{\text{ext}}$ in presence of the external magnetic field B_{ext}. When averaged over the QD ensemble, the field B_f of the FFNS does not contribute to the total magnetic field. However, it causes electron spin relaxation, and hence, decrease in $|\rho_c|$ when B_T approaches zero. Thus, the minimum of $|\rho_c|$ should be observed at $B_{\text{ext}} = -B_N$. This allows us to estimate B_N. The sign of B_N created by light should be opposite for the σ^+- and σ^--polarized excitations. As a result, the minima of $|\rho_c|$ for σ^+- and σ^--polarized excitations are shifted symmetrically from $B_{\text{ext}} = 0$ in opposite directions in Fig. 3.[30] The minima in the two cases differ by $2B_N$. We study the excitation power (P_x) dependence of B_N. A plot of B_N as a function of the scaled excitation power $\rho_x{*}P_x$, where the helicity $\rho_x = +1$ (-1) for σ^+ (σ^-) excitation, shows that the dynamic nuclear polarization builds up linearly with the excitation laser power [Fig. 3(inset)].

We find experimentally that up to $P_x = 50$ mW, B_N remains smaller than 10 mT, and that B_N does not show any indication of saturation. The value of $B_N = 6$ mT observed at $P_x = 50$ mW in our experiments is in agreement with previous reports of B_N measured in an ensemble of InP nano-islands embedded in InGaP matrix.[3] A value of $B_N = 1.2$ T for GaAs QDs formed by interface nano-roughness in a GaAs quantum well has been reported by Gammon *et al.*[1] and Brown *et al.*[31] in single dot measurements, where they estimated that 65% of the nuclear spins were polarized. Yokoi *et al.*[32] have reported $B_N = 160$ mT by using single dot spectroscopy for self-assembled InAlAs QDs, where 6% of the nuclei were polarized. Origin of the small B_N observed for self-assembled InP QD ensemble is not fully clear. We may say that only a small fraction of the nuclei in a QD are polarized, because we do not observe any saturation of B_N up to the highest excitation power used. This may be caused by the low excitation efficiency of the QDs under quasiresonant excitation we used and also by the inefficient transfer of spin polarization from electrons to nuclei.[33] Another possible reason may be the efficient leakage of nuclear spin polarization from the QDs to the surrounding lattice nuclei.[2] Due to the large nuclear spin of In ($I = 9/2$), it may loose its spin polarization rather efficiently through quadrupole interaction in presence of a time varying gradient of local electric field, which may be created by the photogenerated electrons in the QDs.[34] Then, due to close proximity, spin polarization of P nuclei ($I = 1/2$) may be transferred to In nuclei and eventually lost to the surrounding lattice nuclei due to efficient spin relaxation of In nuclei.

D. Electron spin relaxation by slow variation in frozen fluctuations of nuclear spins

For a direct time-domain study of electron spin relaxation we use a pump-probe PL technique.[5,6,15,16] One of the main advantage of this technique is that the measurable time-range of the spin dynamics by this method is not limited by the PL lifetime, in contrast to the time-resolved PL measurements where spin relaxation can be monitored only within the PL decay time.[13] Details of our pump-probe PL experimental arrangement are discussed in Refs. 5,6. In this method, a circularly polarized (σ^+ or σ^-) pump pulse creates a spin orientation of the resident electrons.[18] The spin dynamics is then studied by measuring the decay of ρ_c for the σ^+-polarized probe pulse delayed in time relative to the pump pulse. Pump (probe) power is kept at 1 (0.05) mW, for which $B_N \approx 0$ [Fig. 3(inset)]. Also, the probe power being 20 times smaller than the pump power, it does not destroy the pump-induced spin polarization. A schematic of the pump and probe pulse configurations is shown in the inset of Fig. 4. Exploiting this method we study the spin dynamics in a wide time-range from picoseconds to milliseconds.[5,6]

For the study of spin dynamics in the nanosecond time-regime, pump and probe pulses are derived from a picosecond Ti:Sapphire laser and the pump-probe delay τ

is controlled by optical delay. The polarization-selected PL originating from the probe pulse is time-resolved in a streak camera to monitor the kinetics of ρ_c for the probe-generated PL. Figure 4 shows such kinetics of ρ_c at $\tau = 2$ ns for σ^+-polarized probe pulse when the pump pulse is co-circularly (σ^+) or cross-circularly (σ^-) polarized. Data are taken for $B_{\text{ext}} = 0$ and 0.1 T. As seen in Fig. 4, at times beyond 300 ps, kinetics of ρ_c reaches a constant value (refer to as A_{CP} in Fig. 1), which is strongly negative (positive) for the co- (cross-) circularly polarized pump-probe excitation. The difference ΔA_{CP} between the A_{CP} for the two cases (co- and cross-circularly polarized pump-probe configurations) can be used as a measure of the pump-induced spin polarization of the resident electrons. We find that for a given τ, ΔA_{CP} is sensitive to B_{ext}. The difference $\Delta A_{\text{CP}}(0\text{ T})$ measured at $B_{\text{ext}} = 0$ T is approximately one-third of $\Delta A_{\text{CP}}(0.1\text{ T})$ measured at $B_{\text{ext}} = 0.1$ T for $\tau = 2$ ns [Fig. 4]. An identical behavior is seen when measured at $\tau = 10$ ns. Following the theory of Merkulov *et al.*[10] we conclude that for $B_{\text{ext}} = 0$, a *partial relaxation* (up to one-third of the initial value) of electron spin polarization due to the FFNS takes place within a time shorter than 2 ns. However, the remaining spin polarization does not decay up to 10 ns. In presence of $B_{\text{ext}} = 0.1$ T, electron spin relaxation by the FFNS is suppressed. In that case electron spin relaxation time is much longer than 10 ns.

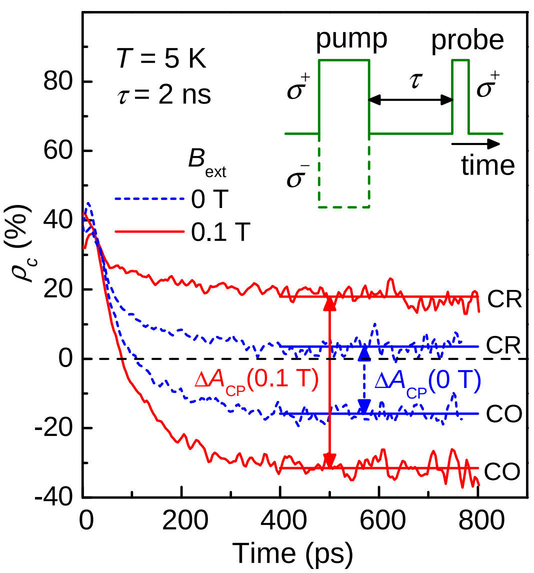

FIG. 4: (Color online) Kinetics of ρ_c for the probe PL at $\tau = 2$ ns for $B_{\text{ext}} = 0$ and 0.1 T. The pump and probe beams are either co-circularly (CO) or cross-circularly (CR) polarized. The difference ΔA_{CP} between A_{CP} for the CR- and CO-cases are indicated by arrows for $B_{\text{ext}} = 0$ and 0.1 T. It is found that $\Delta A_{\text{CP}}(0\text{ T})/\Delta A_{\text{CP}}(0.1\text{ T}) \approx 1/3$. The inset shows a schematic of the pump and probe pulse configurations used in the polarization-resolved pump-probe PL experiment.

As discussed in Sec. I, for $B_{\text{ext}} = 0$ decay of the electron spin polarization surviving after 2 ns (one-third of the initial value) is governed by the NSPE over a longer timescale.[10] For the study of spin dynamics in the microsecond timescale, it is more convenient to measure the magnetic field- and delay-dependence of ρ_c integrated over the PL lifetime. In these experiments, the pump and probe pulses are derived from a CW Ti:Sapphire laser by using acousto-optic modulators (AOMs) which act as electrically controlled gates. Pump and probe pulse widths (1 μs each) and the delay τ between them are controlled by sending electrical pulses to the AOMs from a programable function generator. In this case the accessible range of τ is not limited by the laser pulse repetition period of 12 ns. Details of the experimental setup are discussed in Ref. 6. We measure the difference $\Delta\rho_c$ between ρ_c (integrated over the PL lifetime) for the probe PL for cross- and co-circularly polarized pump-probe excitations.

At first we measure the dependence of $\Delta\rho_c$ on B_{ext} at $\tau = 2$ μs [Fig. 5(a)]. Near $B_{\text{ext}} = 0$ we find that $\Delta\rho_c \approx 0$. This suggests that for $B_{\text{ext}} = 0$, a *total depolarization* of electron spins takes place within 2 μs. This is caused by the NSPE, indicating that the Larmor precession period T_N of nuclear spins in the hyperfine field of electrons is shorter than 2 μs.

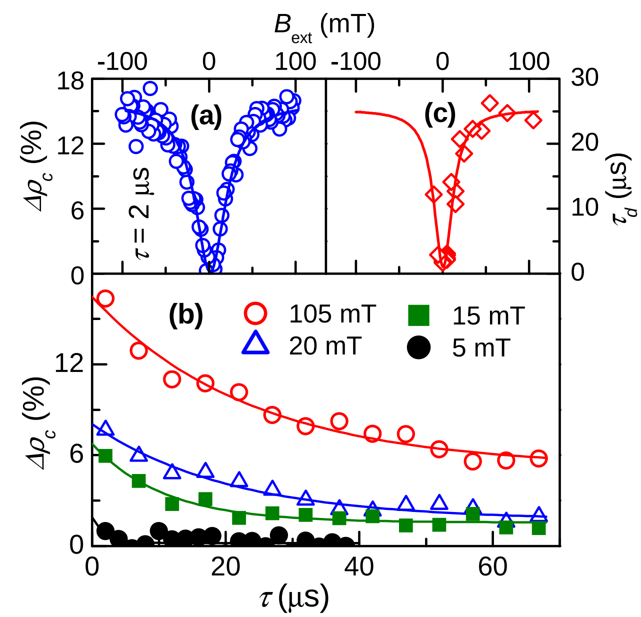

FIG. 5: (Color online) (a) Dependence of $\Delta\rho_c$ on B_{ext} for $\tau = 2$ μs. The solid line is a Lorentzian fit. Total depolarization ($\Delta\rho_c \approx 0$) of the electron spin is seen at $B_{\text{ext}} = 0$. (b) Dependence of $\Delta\rho_c$ on τ at a few values of B_{ext}. Solid lines are exponential fits characterized by the spin decay time τ_d, which is plotted in (c) as a function of B_{ext}. The solid line in (c) is a Lorentzian fit.

To obtain a more quantitative estimate of T_N, we perform a systematic measurement of electron spin polarization decay time τ_d as a function of B_{ext}. Figure 5(b) shows the delay dependence of $\Delta\rho_c$ at different values of B_{ext}. The decay of $\Delta\rho_c$ with τ can be well approximated

by an exponential function, $\Delta\rho_c = A_0 + A_1 \exp(-\tau/\tau_d)$ [Fig. 5(b)]. The decay time τ_d obtained from such fits is shown in Fig. 5(c) for different values of $B_{\rm ext}$. It is seen that τ_d decreases down to 1 μs when $|B_{\rm ext}|$ goes to zero, for which electron spin relaxation takes place due to hyperfine interaction with nuclei.[35] At low temperatures, the *total depolarization* of the electron spins under the vanishing external magnetic field, is caused by the NSPE in the microsecond timescale.[10] Therefore, we assign the time 1 μs obtained from Fig. 5(c) at $B_{\rm ext} = 0$ as an upper limit of T_N.[36] No other experimental study of T_N in QDs is available to our knowledge in the literature for a comparison with our estimate of T_N. However, the value of $T_N \sim 1$ μs obtained by us for the self-assembled InP QDs is comparable to the theoretical estimate of T_N for GaAs QDs.[10] This agreement suggests that T_N may have the same order of magnitude (~ 1 μs) in different self-assembled III-V quantum dots.

IV. CONCLUSION

The effects of nuclear spins on the electron spin dynamics in singly negatively charged InP QDs are studied at low temperature ($T \approx 5$ K), where hyperfine interaction with the nuclear spins is the dominant relaxation channel for the electron spins. We observe that at the vanishing external magnetic field, partial relaxation (up to one-third of the initial value) of electron spin polarization takes place within ~ 1 ns due to the frozen fluctuations of the nuclear hyperfine field. A value of 15 mT is estimated for the effective magnetic field B_f of the frozen fluctuations of nuclear spins. Total depolarization of electron spins with a characteristic time of 1 μs is observed at the vanishing external magnetic field, due to the slow variation of B_f in time caused by the nuclear spin precession in the hyperfine field of electrons. The characteristic time of 1 μs is assigned to the nuclear spin precession period T_N in the hyperfine field of electrons. At high excitation power the dynamic nuclear polarization is observed, giving rise to an Overhauser field $B_N = 6$ mT at 50 mW excitation.

Acknowledgments

Authors thank V. K. Kalevich and I. Ya. Gerlovin for fruitful discussions. The work is partially supported by "Grant-in-Aid for Scientific Research" Nos. 17-5056, 13852003, and 18204028 from the MEXT of Japan and "R&D Promotion Scheme Funding International Joint Research" promoted by NICT of Japan, by ISTC, grant 2679, by Russian Ministry of Sci. & Edu., grant RNP.2.1.1.362, and by RFBR, grant 06-02-17137-a.

[*] E-mail: bipulpal@sakura.cc.tsukuba.ac.jp
[†] E-mail: syuv54@mail.ru

[1] D. Gammon, Al. L. Efros, T. A. Kennedy, M. Rosen, D. S. Katzer, D. Park, S. W. Brown, V. L. Korenev, and I. A. Merkulov, Phys. Rev. Lett. **86**, 5176 (2001).
[2] *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya, Modern Problems in Condensed Matter Sciences Vol. 8 (North-Holland, Amsterdam, 1984).
[3] R. I. Dzhioev, B. P. Zakharchenya, V. L. Korenev, P. E. Pak, M. N. Tkachuk, D. A. Vinokurov, and I. S. Tarasov, Pis'ma Zh. Eksp. Teor. Fiz. **68**, 711 (1998) [JETP Lett. **68**, 745 (1998)].
[4] R. I. Dzhioev, B. P. Zakharchenya, V. L. Korenev, and M. V. Lazarev, Fiz. Tverd. Tela (St. Petersburg) **41**, 2193 (1999) [Phys. Solid State **41**, 2014 (1999)].
[5] M. Ikezawa, B. Pal, Y. Masumoto, I. V. Ignatiev, S. Yu. Verbin, and I. Ya. Gerlovin, Phys. Rev. B **72**, 153302 (2005).
[6] B. Pal, M. Ikezawa, Y. Masumoto, and I. V. Ignatiev, J. Phys. Soc. Jpn. **75**, 054702 (2006).
[7] *Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth, Nanoscience and Technology (Springer, Berlin, 2002).
[8] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[9] V. L. Berkovits, A. I. Ekimov, and V. I. Safarov, Zh. Eksp. Teor. Fiz. **65**, 346 (1973) [Sov. Phys. JETP **38**, 169 (1974)].
[10] I. A. Merkulov, Al. L. Efros, and M. Rosen, Phys. Rev. B **65**, 205309 (2002).
[11] A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. **88**, 186802 (2002).
[12] Y. G. Semenov and K. W. Kim, Phys. Rev. B **67**, 073301 (2003).
[13] P.-F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K. V. Kavokin, O. Krebs, P. Voisin, and Y. Masumoto, Phys. Rev. Lett. **94**, 116601 (2005).
[14] See e.g., Fig. 3 of Ref. 13.
[15] S. Cortez, O. Krebs, S. Laurent, M. Senes, X. Marie, P. Voisin, R. Ferreira, G. Bastard, J.-M. Gérard, and T. Amand, Phys. Rev. Lett. **89**, 207401 (2002).
[16] J. S. Colton, T. A. Kennedy, A. S. Bracker, and D. Gammon, Phys. Rev. B **69**, 121307(R) (2004).
[17] I. E. Kozin, V. G. Davydov, I. V. Ignatiev, A. V. Kavokin, K. V. Kavokin, G. Malpuech, H.-W. Ren, M. Sugisaki, S. Sugou, and Y. Masumoto, Phys. Rev. B **65**, 241312(R) (2002).
[18] A model of optical orientation of resident electron spins for *quasiresonant* excitation in the excited state of a singly negatively charged QDs, applicable for our experimental condition, was discussed in details in Ref. 6.
[19] Y. Masumoto, S. Oguchi, B. Pal, and M. Ikezawa, Phys. Rev. B **74**, 205332 (2006).
[20] Recently, negative degree of circular polarization of PL from the negatively charged QDs has been reported by other researchers, see e.g., Refs. 15,21,22,23. Unlike the *quasiresonant* excitation in the QD excited state used in our experiments, measurements of Refs. 15,21,22,23 have been performed under wetting layer excitation and the holes were assumed to be *totaly depolarized* before cap-

turing in the QDs.
[21] S. Laurent, O. Krebs, S. Cortez, M. Sénès, X. Marie, T. Amand, P. Voisin, and J.-M. Gérard, Physica E **20**, 404 (2004).
[22] V. K. Kalevich, I. A. Merkulov, A. Y. Shiryaev, K. V. Kavokin, M. Ikezawa, T. Okuno, P. N. Brunkov, A. E. Zhukov, V. M. Ustinov, and Y. Masumoto, Phys. Rev. B **72**, 045325 (2005).
[23] S. Laurent, M. Sénès, O. Krebs, V. K. Kalevich, B. Urbaszek, X. Marie, T. Amand, and P. Voisin, Phys. Rev. B **73**, 235302 (2006).
[24] This behavior is similar to that observed for optical pumping of electron spins in n-type semiconductors.[2] We estimate that the number of photocreated electron-hole pair per QD per pulse remains less than one up to the highest average excitation power (50 mW) used.
[25] M. E. Ware, E. A. Stinaff, D. Gammon, M. F. Doty, A. S. Bracker, D. Gershoni, V. L. Korenev, S. C. Badescu, Y. Lyanda-Geller, and T. L. Reinecke, Phys. Rev. Lett. **95**, 177403 (2005).
[26] K. V. Kavokin, Phys. Status Solidi (a) **195**, 592 (2003).
[27] T. Flissikowski, I. A. Akimov, A. Hundt, and F. Henneberger, Phys. Rev. B **68**, 161309(R) (2001).
[28] S. Laurent, B. Eble, O. Krebs, A. Lemaître, B. Urbaszek, X. Marie, T. Amand, and P. Voisin, Phys. Rev. Lett. **94**, 147401 (2005).
[29] Y. Masumoto, I. V. Ignatiev, K. Nishibayashi, T. Okuno, S. Yu. Verbin, and I. A. Yugova, J. Lumin. **108**, 177 (2004).
[30] The minimum of $|\rho_c|$ appears at $B_{\rm ext} \approx 0$ when the polarization of the excitation beam is modulated between σ^+ and σ^- at a frequency of 42 kHz, by using a photo-elastic modulator in the excitation path. It is well known that in such a case dynamic nuclear polarization cannot build up.[2]
[31] S. W. Brown, T. A. Kennedy, D. Gammon, and E. S. Snow, Phys. Rev. B **54**, R17339 (1996).
[32] T. Yokoi, S. Adachi, H. Sasakura, S. Muto, H. Z. Song, T. Usuki, and S. Hirose, Phys. Rev. B **71**, 041307(R) (2005).
[33] A. Imamoglu, E. Knill, L. Tian, and P. Zoller, Phys. Rev. Lett. **91**, 017402 (2003).
[34] G. Salis, D. D. Awschalom, Y. Ohno, and H. Ohno, Phys. Rev. B **64**, 195304 (2001).
[35] For $B_{\rm ext} \neq 0$, the effect of hyperfine interaction on the electron spin relaxation begins to diminish, because the external magnetic field starts to influence the electron-nuclear spin dynamics. Electron spin decay time τ_d measured for $|B_{\rm ext}| > 0$ cannot be directly assigned to the nuclear spin precession period T_N in the hyperfine field of electrons. Effects of the external magnetic field on the electron-nuclear spin dynamics is discussed in Ref. 10.
[36] Accuracy of this estimate is decided by the accuracy of the exponential fit, estimated at 30% in Fig. 5(b).